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Examiner : Carlos N. Lopez  
Art Unit : 1731  
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**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

Applicants : J. SUGAWARA et al.  
Serial No. : 10/019,752  
Filed : December 27, 2001  
For : BLACK CERAMIC SINTER WITH LOW THERMAL EXPANSION  
AND HIGH SPECIFIC RIGIDITY AND PROCESS FOR  
PRODUCING THE SAME

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

**SUBMISSION OF TRANSLATION OF PRIORITY DOCUMENT**

SIR:

The above-identified patent application claims foreign priority under 35 U.S.C. §119 from Japanese Patent Application No. 11-183109 filed June 29, 1999.

The Patent and Trademark Office has acknowledged the claim for foreign priority under 35 U.S.C. §119 and has acknowledged a copy of a certified copy of the priority document has been received in this national stage application from the International Bureau.

In order to complete the claim for foreign priority under 35 U.S.C. §119, submitted herewith is an English translation, under Declaration, or priority Japanese Patent Application No. 11-183109 filed June 29, 1999.

Please charge any required fee associated with this submission to Deposit Account No. 11-0600. A duplicate of this paper is enclosed for deposit account charging purposes.

Respectfully submitted,

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In the matter  
of U.S. Patent Application  
No. 10/019,752  
of Nippon Steel Corporation

D E C L A R A T I O N

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do solemnly and sincerely declare:

1. That I am well acquainted with the English and  
Japanese languages, and
2. That the attached document is a full and faithful  
translation into the English language made by me of  
Japanese Patent Application No. 11-183109 duly certified by  
Commissioner of Patent Office to the best of my knowledge  
and belief.

Declared at Tokyo Japan on this 4th day of October, 2005.

Masayo Yamauchi  
Masayo YAMAUCHI

JAPAN PATENT OFFICE

This is to certify that the annexed is a true copy of the following application as filed with this office.

Date of Application: June 29, 1999

Application Number: 11-183109

Applicant(s): Nippon Steel Corporation

Commissioner, Japan Patent Office

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[DOCUMENT'S NAME] SPECIFICATION

[TITLE] BLACK LOW THERMAL EXPANSION CERAMIC SINTERED  
BODY AND METHOD FOR PRODUCTION THEREOF

[SCOPE OF CLAIM FOR A PATENT]

5 [Claim 1] A black low thermal expansion high specific  
rigidity ceramic sintered body, characterized by having a  
thermal expansion coefficient of not more than  $0.6 \times 10^{-6}/^{\circ}\text{C}$   
in absolute value at room temperature, a modulus of elasticity  
(Young's modulus) of not less than 100 GPa, and specific  
10 rigidity (Young's modulus/specific gravity) of not less than  
40  $\text{GPa} \cdot \text{cm}^3/\text{g}$  and assuming a black tone.

[Claim 2] A black low thermal expansion high specific  
rigidity ceramic sintered body, characterized by having a  
chemical composition comprising 8.0 - 17.2 mass % of MgO,  
15 22.0 - 38.0 mass % of  $\text{Al}_2\text{O}_3$ , 49.5 - 65.0 mass % of  $\text{SiO}_2$ , a total  
of 0.1 - 2 mass % of one or more transition elements as reduced  
to oxides, and 0 - 2.5 mass % of  $\text{Li}_2\text{O}$ , and having the mass  
ratios satisfy the relationships of  $(\text{SiO}_2 - 8 \times \text{Li}_2\text{O})/\text{MgO} \geq 3.0$  and  $(\text{SiO}_2 - 8 \times \text{Li}_2\text{O})/\text{Al}_2\text{O}_3 \geq 1.2$ .

20 [Claim 3] A black low thermal expansion high specific  
rigidity ceramic sintered body according to claim 2, wherein  
the thermal expansion coefficient is not more than  $0.6 \times 10^{-6}/^{\circ}\text{C}$   
in absolute value at room temperature, the modulus of  
elasticity (Young's modulus) is not less than 100 GPa, and  
25 the specific rigidity (Young's modulus/specific gravity) is  
not less than 40  $\text{GPa} \cdot \text{cm}^3/\text{g}$  and the tone of the sintered body  
is black.

[Claim 4] A black low thermal expansion high specific  
rigidity ceramic sintered body according to any one of claims  
30 1 through 3, wherein the total reflectivity of the sintered  
body is not more than 17% at a wavelength of light in the  
range of 200 - 950 nm.

[Claim 5] A black low thermal expansion high specific rigidity ceramic sintered body according to any one of claims 1 through 4, wherein the apparent porosity of the sintered body is not more than 2%.

5 [Claim 6] A black low thermal expansion high specific rigidity ceramic sintered body according to any one of claims 1 through 5, wherein not less than 80 vol. % of the crystal phase of the sintered body is a crystal phase of cordierite.

[Claim 7] A black low thermal expansion high specific  
10 rigidity ceramic sintered body according to any one of claims 1, 3, 4, 5, and 6, wherein the thermal expansion coefficient is not more than  $0.3 \times 10^{-6}/^{\circ}\text{C}$  in absolute value at room temperature.

[Claim 8] A black low thermal expansion high specific  
15 rigidity ceramic sintered body according to any one of claims 1, 3, 4, 5, 6, and 7, wherein the modulus of elasticity is not less than 120 GPa and the specific rigidity is not less than  $50 \text{ GPa} \cdot \text{cm}^3/\text{g}$ .

[Claim 9] A black low thermal expansion high specific  
20 rigidity ceramic sintered body according to any one of claims 2 through 8, wherein the chemical composition has such mass ratios as satisfy the relationships of  $(\text{SiO}_2 - 8 \times \text{Li}_2\text{O})/\text{MgO} \geq 3.65$  and  $(\text{SiO}_2 - 8 \times \text{Li}_2\text{O})/\text{Al}_2\text{O}_3 \geq 1.3$ .

[Claim 10] A method for the production of a black low thermal  
25 expansion high specific rigidity ceramic sintered body, characterized by forming the sintered body in an atmosphere of a non-oxidizing gas at a temperature in the range of  $1200 - 1500^{\circ}\text{C}$ .

[Claim 11] A method for the production of a black low thermal  
30 expansion high specific rigidity ceramic sintered body set forth in any one of claims 1 through 9, wherein the sintered body is formed in an atmosphere of a non-oxidizing gas at



a temperature in the range of 1200 - 1500°C.

[Claim 12] A method for the production of a black low thermal expansion high specific rigidity ceramic sintered body according to claim 10 or 11, wherein the non-oxidizing gas  
5 is one or more members selected among argon, helium, nitrogen and hydrogen.

[Claim 13] A method for the production of a black low thermal expansion high specific rigidity ceramic sintered body according to any one of claims 10 through 12, wherein the  
10 raw material powder is one or more members selected from the group consisting of cordierite powder, talc, magnesia spinel, magnesia, magnesium hydroxide, magnesium carbonate,  $\text{Li}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2$  type powders (petalite, spodumene, and eucryptite), lithium hydroxide, lithium carbonate, alumina  
15 powder, silica powder, kaolin powder, and mullite powder.

[Claim 14] A method for the production of a black low thermal expansion high specific rigidity ceramic sintered body according to claim 13, wherein not less than 70 mass % of the MgO component as the MgO-source raw material is supplied  
20 by one or more members selected from the group consisting of electro-molten cordierite powder, synthetic cordierite powder, and talc powder.

[Claim 15] A method for the production of a black low thermal expansion high specific rigidity ceramic sintered body according to any one of claims 10 through 14, wherein the sintering method is a hot press method, an HIP method, a gas pressure sintering method, or a normal pressure sintering method.  
25

#### [DETAILED DESCRIPTION OF THE INVENTION]

30 [0001]

[Technical Filed of the Invention]

This invention relates to a black low thermal expansion

ceramic sintered body possessing a black tone, manifesting very small thermal expansion at room temperature, and abounding in rigidity and specific rigidity, and to a method for the production thereof.

5 [0002]

[Prior Art]

Recently, owing to the trend of semiconductors toward higher integration and magnetic heads toward further miniaturization, the production units (such as an exposure  
10 meter, a processing machine, and a construction machine) for such semiconductors and magnetic heads, measuring devices, measuring prototypes, and reflecting mirrors have reached the point of requiring high dimensional accuracy and high rigidity. For these devices, the stability of dimensional  
15 accuracy has also come to gain in significance. The prevention of such devices from incurring the deformation which is caused by the fluctuation of an ambient temperature or the emission of heat from the device itself has become an important task. The materials which produce very small thermal expansions  
20 and abound in rigidity and specific rigidity (Young's modulus/specific gravity) have come to find the use for component members in such devices.

[0003]

The various devices as mentioned above are mostly aimed  
25 at handling the lights such as laser beam, ultraviolet light, and visible ray for the purpose of exposure or measurement. The members which are used in these devices more often than not abhor unnecessary reflection (including irregular reflection) or transmission of light. On many occasions,  
30 black materials which succumb only sparingly to reflection and transmission of light are found as necessary supplies.

[0004]

The low thermal expansion materials which have heretofore known in the art include Invar alloy (Fe-Ni type) and super-Invar alloy (Fe-Ni-Co type) in the class of metals, such low thermal expansion glasses as ZERODUR(TM) glass  
5 (available from Schott ML GmbH in Germany), quartz glass ( $\text{SiO}_2$ ), and titanium dioxide-containing quartz glass ( $\text{SiO}_2\text{-TiO}_2$ ) in the class of ceramics, aluminum titanate ( $\text{TiO}_2\text{-Al}_2\text{O}_3$ ), cordierite ( $\text{MgO-Al}_2\text{O}_3\text{-SiO}_2$ ) type sintered body and glass, lithium-alumino-silicate ( $\text{Li}_2\text{O-Al}_2\text{O}_3\text{-SiO}_2$ ) type sintered body  
10 and glass. However, among those low thermal expansion materials, examples of the materials in which the materials themselves have made to possess a black tone have not known.

[0005]

The super-Invar alloy as a low thermal expansion metal  
15 indeed manifests such a low thermal expansion coefficient as  $0.13 \times 10^{-6}/^\circ\text{C}$  at room temperature and yet has such a large specific gravity as  $8.2 \text{ g/cm}^3$  and such a not very high Young's modulus as 125 GPa. Thus, it has a very low specific rigidity approximately of  $15 \text{ GPa}\cdot\text{cm}^3/\text{g}$  and, therefore, is deficient  
20 in mechanical stability. The term "specific rigidity" as used herein means a magnitude obtained by dividing a Young's modulus (E) by a specific gravity ( $\rho$ ) (specific rigidity =  $E/\rho$ ). The impartation of the black color to the surface of this alloy has no alternative but to rely on the method of using such  
25 a surface coat as a black Cr plating on the surface layer. The use of the surface coat, however, entails the problem of exerting adverse effects on the low thermal expansion property and the precision machining property.

[0006]

30 The quartz glass, while enjoying such a low thermal expansion coefficient as  $0.48 \times 10^{-6}/^\circ\text{C}$ , suffers from such an insufficient specific rigidity as about  $33 \text{ GPa}\cdot\text{cm}^3/\text{g}$  and a

clear tone.

[0007]

The ZERODUR(TM) glass has been finding the extensive utility in applications to such devices as measuring  
5 prototypes. It enjoys such a sufficiently low thermal expansion coefficient as  $0.02 \times 10^{-6}/^{\circ}\text{C}$  at room temperature and yet suffers from a clear tone. It further encounters difficulty in forming products too complicate in shape and products too large in size to manufacture. Further, since  
10 it manifests specific rigidity and Young's modulus respectively approximating  $35.6 \text{ GPa} \cdot \text{cm}^3/\text{g}$  and  $90 \text{ GPa}$ , it does not fully fit the use aimed at by this invention.

[0008]

As regards the aluminum titanate, it has been known to  
15 have produced a sintered body manifesting such a low thermal expansion coefficient as  $-0.068 \times 10^{-6}/^{\circ}\text{C}$  ("Glossary of Fine Ceramics Catalogs (1987)", p. 140). This compound manifests such a high water absorption as 1.59% and thus offers only insufficient denseness for the use aimed at by this invention.  
20 No sintered bodies of this compound has been known to possess a black tone.

[0009]

The lithium-alumino-silicate type sintered body and glass are deficient in mechanical stability because it  
25 manifests only such insufficiently high specific rigidity as not more than  $33 \text{ GPa} \cdot \text{cm}^3/\text{g}$  in spite of such a small thermal expansion coefficient as falls in the range of  $-5$  to  $1 \times 10^{-6}/^{\circ}\text{C}$ . It has predominantly acquired a white tone and has not acquired a black tone so far.

30 [0010]

A cordierite type sintered body has been known as a sintered body manifesting a low thermal expansion coefficient

and a relatively high specific rigidity. The cordierite type sintered body has a low thermal expansion coefficient of not more than  $0.6 \times 10^{-6}/^{\circ}\text{C}$  at room temperature, and a relatively high specific rigidity. However, a cordierite sintered body  
5 acquired a black tone has not been reported.

[0011]

JP-A-61-72,679 discloses a low thermal expansion ceramic sintered body which has a chemical composition mainly comprising 0.3 - 5.5 mass % of  $\text{Li}_2\text{O}$ , 4.1 - 16.4 mass % of  $\text{MgO}$ ,  
10 20.7 - 42.8 mass % of  $\text{Al}_2\text{O}_3$ , and 46.3 - 70.1 mass % of  $\text{SiO}_2$ , a crystal phase containing not less than 30 mass % of cordierite and not less than 5 mass % of  $\beta$ -spodumene as main components, and manifests a thermal expansion coefficient of  $2.0 \times 10^{-6}/^{\circ}\text{C}$  at a temperature in the range of  $20^{\circ}\text{C}$  -  $800^{\circ}\text{C}$ . This publication,  
15 however, has absolutely no mention of the tone of the sintered body and points out the fact that the sintered body produced by the method taught in the publication does not acquire a black tone (refer to Comparative Example 22 in Table 1 inserted in the working example which will be specifically described  
20 hereinbelow).

[0012]

JP-A-10-53,460 reports a dense ceramic substance which comprises 1.5 - 6.5 mass % of  $\text{Li}_2\text{O}$ , 1.0 - 10 mass % of  $\text{MgO}$ , 14 - 30 mass % of  $\text{Al}_2\text{O}_3$ , and 58 - 83 mass % of  $\text{SiO}_2$ , and allows  
25 the coexistence of petalite, spodumene, and cordierite in a crystal phase and demonstrates that this substance excels in resistance to thermal shock. This publication, however, has absolutely no mention of the tone of the ceramic and points out the fact that the sintered body produced by the method  
30 taught in the publication does not acquire a black tone (refer to Comparative Example 23 in Table 1 inserted in the working example which will be specifically described hereinbelow).

[0013]

"Ceramics", Vol. 18 (1983) No. 5 discloses a Co-Cr-Fe type spinel, a Co-Mn-Fe type spinel, a Co-Mn-Cr-Fe type spinel, a Co-Ni-Cr-Fe type spinel, and a Co-Ni-Mn-Cr-Fe type spinel as black pigments for the use in coloring ceramics and also discloses a solid solution of Sb in  $\text{SiO}_2$  and a solid solution of Co and Ni in  $\text{ZrSiO}_4$  as gray pigments. These pigments, however, are intended to utilize the phenomenon of coloration in the glaze on the surface of ceramics and not to impart a black color to a depth in the sintered body itself. Any attempt to use the glaze on the surface of a low thermal expansion ceramic substance proves futile because the difference in thermal expansion between the ceramic substance and the glaze tends to inflict a crack to the applied layer of the glaze.

[0014]

The silicon carbide sintered body has been commercially available as a black ceramic substance. JP-A-08-310,860 discloses a black zirconia ceramic sintered body, JP-A-04-50,160 a method for the production of a high rigidity black alumina sintered body, and JP-A-06-172,034 a black silicon nitride sintered body, respectively. Though these sintered bodies are black, their thermal expansion coefficients at room temperature are  $2.3 \times 10^{-6}/^\circ\text{C}$  in the sintered body of silicon carbide,  $7 \times 10^{-6}/^\circ\text{C}$  in that of zirconia,  $5.3 \times 10^{-6}/^\circ\text{C}$  in that of alumina, and  $1.3 \times 10^{-6}/^\circ\text{C}$  in that of silicon nitride. Thus, these sintered bodies are incapable of realizing a low thermal expansion coefficient aimed at this invention.

[0015]

Incidentally, the term "room temperature" as used in this invention refers to the range of temperatures,  $20^\circ\text{C}$  -

25°C. The room temperature mentioned in the present specification invariably refers to this temperature range.

[0016]

JP-B-57-29,436 discloses a technique which comprises  
5 adding to a cordierite sintered body an oxide of such a transition element as Zn, V, Nb, Cr, Mo, or W for the purpose of densifying the sintered body. The sintered body obtained by this technique, however, manifests such an insufficiently low thermal expansion coefficient as  $0.96 \times 10^{-6}/^{\circ}\text{C}$ , fails to  
10 acquire sufficient densification as evident from water absorption of 4.6%, and suffers from not sufficiently high rigidity. The publication has absolutely no mention of the tone.

[0017]

15 [Problem to be Solved by the Invention]

The material which assumes a black color, manifests low thermal expansion, and possesses rigidity and specific rigidity high enough to ensure effective use as building materials has not been known to date.

20 [0018]

This invention is aimed at providing a black low thermal expansion ceramic sintered body which assumes a black tone and manifests very low thermal expansion and high rigidity and specific rigidity at room temperature and a method for  
25 the production thereof.

[0019]

[Means for Solving Problem]

To be specific, this invention is featured by the following items.

30 (1) A black low thermal expansion high specific rigidity ceramic sintered body, characterized by having a thermal expansion coefficient of not more than  $0.6 \times 10^{-6}/^{\circ}\text{C}$  in absolute

value at room temperature, a modulus of elasticity (Young's modulus) of not less than 100 GPa, and specific rigidity (Young's modulus/specific gravity) of not less than 40 GPa·cm<sup>3</sup>/g and assuming a black tone.

5       (2) A black low thermal expansion high specific rigidity ceramic sintered body, characterized by having a chemical composition comprising 8.0 - 17.2 mass % of MgO, 22.0 - 38.0 mass % of Al<sub>2</sub>O<sub>3</sub>, 49.5 - 65.0 mass % of SiO<sub>2</sub>, a total of 0.1 - 2 mass % of one or more transition elements as reduced to  
10 oxides, and 0 - 2.5 mass % of Li<sub>2</sub>O, and having the mass ratios satisfy the relationships of  $(\text{SiO}_2 - 8 \times \text{Li}_2\text{O})/\text{MgO} \geq 3.0$  and  $(\text{SiO}_2 - 8 \times \text{Li}_2\text{O})/\text{Al}_2\text{O}_3 \geq 1.2$ .

      (3) A black low thermal expansion high specific rigidity ceramic sintered body according to the item (2), wherein the  
15 thermal expansion coefficient is not more than  $0.6 \times 10^{-6}/^\circ\text{C}$  in absolute value at room temperature, the modulus of elasticity (Young's modulus) is not less than 100 GPa, and the specific rigidity (Young's modulus/specific gravity) is not less than 40 GPa·cm<sup>3</sup>/g and the tone of the sintered body  
20 is black.

      (4) A black low thermal expansion high specific rigidity ceramic sintered body according to any one of the items (1) through (3), wherein the total reflectivity of the sintered body is not more than 17% at a wavelength of light in the  
25 range of 200 - 950 nm.

      (5) A black low thermal expansion high specific rigidity ceramic sintered body according to any one of the items (1) through (4), wherein the apparent porosity of the sintered body is not more than 2%.

30       (6) A black low thermal expansion high specific rigidity ceramic sintered body according to any one of the items (1) through (5), wherein not less than 80 vol. % of the crystal



phase of the sintered body is a crystal phase of cordierite.

(7) A black low thermal expansion high specific rigidity ceramic sintered body according to any one of the items (1) and (3) through (6), wherein the thermal expansion coefficient is not more than  $0.3 \times 10^{-6}/^{\circ}\text{C}$  in absolute value at room temperature.

(8) A black low thermal expansion high specific rigidity ceramic sintered body according to any one of the items (1) and (3) through (7), wherein the modulus of elasticity is not less than 120 GPa and the specific rigidity is not less than  $50 \text{ GPa} \cdot \text{cm}^3/\text{g}$ .

(9) A black low thermal expansion high specific rigidity ceramic sintered body according to any one of the items (2) through (8), wherein the chemical composition has such mass ratios as satisfy the relationships of  $(\text{SiO}_2 - 8 \times \text{Li}_2\text{O})/\text{MgO} \geq 3.65$  and  $(\text{SiO}_2 - 8 \times \text{Li}_2\text{O})/\text{Al}_2\text{O}_3 \geq 1.3$ .

(10) A method for the production of a black low thermal expansion high specific rigidity ceramic sintered body, characterized by forming the sintered body in an atmosphere of a non-oxidizing gas at a temperature in the range of  $1200 - 1500^{\circ}\text{C}$ .

(11) A method for the production of a black low thermal expansion high specific rigidity ceramic sintered body set forth in any one of the items (2) through (9), wherein the sintered body is formed in an atmosphere of a non-oxidizing gas at a temperature in the range of  $1200 - 1500^{\circ}\text{C}$ .

(12) A method for the production of a black low thermal expansion high specific rigidity ceramic sintered body according to the item (10) or (11), wherein the non-oxidizing gas is one or more members selected among argon, helium, nitrogen and hydrogen.

(13) A method for the production of a black low thermal

expansion high specific rigidity ceramic sintered body according to any one of the items (10) through (12), wherein the raw material powder is one or more members selected from the group consisting of cordierite powder, talc, magnesia  
5 spinel, magnesia, magnesium hydroxide, magnesium carbonate,  $\text{Li}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2$  type powders (petalite, spodumene, and eucryptite), lithium hydroxide, lithium carbonate, alumina powder, silica powder, kaolin powder, and mullite powder.

(14) A method for the production of a black low thermal  
10 expansion high specific rigidity ceramic sintered body according to the item (13), wherein not less than 70 mass % of the MgO component as the MgO-source raw material is supplied by one or more members selected from the group consisting of electro-molten cordierite powder, synthetic cordierite  
15 powder, and talc powder.

(15) A method for the production of a black low thermal expansion high specific rigidity ceramic sintered body according to any one of the items (10) through (14), wherein the sintering method is a hot press method, an HIP method,  
20 a gas pressure sintering method, or a normal pressure sintering method.

[0020]

The sintered body of this invention can, by having a chemical composition mainly comprise 8.0 - 17.2 mass % of  
25 MgO, 22.0 - 38.0 mass % of  $\text{Al}_2\text{O}_3$ , and 49.5 - 65.0 mass % of  $\text{SiO}_2$  and consequently forming a crystal composition mainly as a cordierite crystal phase, obtain a low thermal expansion coefficient and a high specific rigidity which are aimed at by this invention.

30 [0021]

When the acquisition of the low thermal expansion coefficient at room temperature is not aimed at as in the

present invention,  $\text{Li}_2\text{O}$  does not constitute itself an essential component. The incorporation of  $\text{Li}_2\text{O}$ , however, may bring about such effects as enhancing the sintering properties, facilitating the formation of a dense sintered body, allowing  
5 the activation of the reaction (mass transfer) in the sintered body in the process of formation to start at a relatively low temperature, enabling the impartation of a black color to start at a still lower temperature, and causing the finally produced sintered body to be further blackened.

10 [0022]

The sintered body of this invention can, by incorporating a total of 0.1 - 2 mass % of one or more transition elements as reduced to oxides, having the mass ratios in a chemical composition satisfy the relationship:  $X = (\text{SiO}_2 - 8 \times \text{Li}_2\text{O}) / \text{MgO}$   
15  $\geq 3.0$  and  $Y = (\text{SiO}_2 - 8 \times \text{Li}_2\text{O}) / \text{Al}_2\text{O}_3 \geq 1.2$ , and effecting the sintering process in an atmosphere of a non-oxidizing gas at a temperature in the range of 1200 - 1500°C, acquire a black tone which is aimed at by this invention.

[0023]

20 [Mode for Carrying Out the Invention]

For the quantitative determination about whether a given sintered body has a black tone or not, a method which relies on the color difference found with a colorimeter to attain the expected expression or a method which utilizes the total  
25 reflectivity specified in JIS K7105 may be adopted. The applications to be found for the sintered body of this invention are such that they abhor the reflection of light including irregular reflection and, therefore, are most appropriately rated for the degree of this abhorrence by utilizing the total  
30 reflectivity. The total reflectivity is attained by combined determination of direct reflection and diffused reflection with the aid of a globular integrating sphere. The

determination in this case is made in accordance with JIS K7105.

[0024]

Generally, for such members used in devices handling  
5 a laser beam or an ultraviolet light and requiring a black  
tone, products plated with black chrome and products treated  
with black alumite may be adopted. The total reflectivity  
in the products plated with black chrome is in the range of  
5 - 7% at a wavelength of light in the range of 200 - 950  
10 nm and in the products treated with black alumite in the range  
of 6 - 8% at a wavelength of light in the range of 200 - 650  
nm or in the range of 10 - 60% at a wavelength of light in  
the range of 700 - 950 nm. The total reflectivity is preferred  
to be as low as permissible because the degree with which  
15 the reflection is prevented is heightened in proportion as  
the total reflectivity is lowered. Generally, so long as the  
total reflectivity is not more than 17% within the range,  
200 - 950 nm, of wavelength of light, members for such devices  
as abhorring the reflection of light can be safely used from  
20 the practical point of view. Better results can be obtained  
when the total reflectivity is not more than 12%.

[0025]

In this invention, the sintered body of a black tone  
which is aimed at by this invention is characterized by having  
25 incorporated therein as a coloring auxiliary one or more  
transition elements in a total amount of 0.1 - 2 mass % as  
reduced to oxides, having the mass ratios of the chemical  
composition satisfy the relationship:  $X = (\text{SiO}_2 - 8 \times \text{Li}_2\text{O}) / \text{MgO} \geq 3.0$  and  $Y = (\text{SiO}_2 - 8 \times \text{Li}_2\text{O}) / \text{Al}_2\text{O}_3 \geq 1.2$ , and performing  
30 the sintering process in an atmosphere of a non-oxidizing  
gas at a temperature in the range of 1200 - 1500°C. The  
fulfillment of all these requirements enables the total

reflectivity of the sintered body to fall in the range contemplated by this invention.

[0026]

No fully satisfactory black tone can be obtained when  
5 the total amount of transition elements as coloring auxiliary  
is not more than 0.1 mass % as reduced to oxides. If the total  
amount is not less than 2 mass %, the excess would be at a  
disadvantage in compelling the produced sintered body to form  
10 a low melting compound therein and giving rise to a phenomenon  
of foaming and bringing about the degradation of density and  
rigidity. The total amount of transition elements as reduced  
to oxides is more preferably in the range of 0.3 - 1 mass %.  
By having the total amount fall in this range, it is made  
possible to obtain a sintered body having sufficient blackness  
15 and abounding in density and rigidity.

[0027]

As the transition elements which are usable in this  
invention, such first-row transition elements as Cr, Mn, Fe,  
Co, Ni, and Cu prove most advantageous among other transition  
20 elements.

[0028]

When the amount of  $\text{SiO}_2$  decreases in the mass percentage  
composition,  $\text{MgO-Al}_2\text{O}_3\text{-SiO}_2$ , as the main component of the  
sintered body, the black color imparted to the sintered body  
25 is proportionately lightened. The  $\text{Li}_2\text{O}$  included in the  
composition, while the composition is in the process of forming  
a sintered body, fixes a part of  $\text{SiO}_2$  in the form of a  
 $\text{Li}_2\text{O-Al}_2\text{O}_3\text{-SiO}_2$  sintered body. When the percentage  
composition is considered, therefore, it requires subtraction  
30 of that part of the amount of  $\text{SiO}_2$  from the  $\text{SiO}_2$  amount.  
Empirically, it is inferred that the amount of  $\text{SiO}_2$  so  
subtracted is approximately represented by  $8 \times \text{Li}_2\text{O}$ .

Accordingly, for the purpose of obtaining the black coloration contemplated by this invention, it is necessary to transform the mass ratio in the chemical composition to this composition ratio,  $X = (\text{SiO}_2 - 8 \times \text{Li}_2\text{O}) / \text{MgO} \geq 3.0$  and  $Y = (\text{SiO}_2 - 8 \times \text{Li}_2\text{O}) / \text{Al}_2\text{O}_3 \geq 1.2$ , preferably  $X = (\text{SiO}_2 - 8 \times \text{Li}_2\text{O}) / \text{MgO} \geq 3.65$  and  $Y = (\text{SiO}_2 - 8 \times \text{Li}_2\text{O}) / \text{Al}_2\text{O}_3 \geq 1.3$ . The mechanism responsible for imparting a fully satisfactory black tone to a sintered body in consequence of securing the composition ratio of  $\text{SiO}_2$  as described above has not yet been fully elucidated. It may be logically explained, however, by a postulate that the condition of the deficiency of oxygen in the Si-O bond present in a crystal constitutes itself a factor of some sort or other.

[0029]

For the sake of imparting a black tone to a given sintered body, the sintering atmosphere and the sintering temperature adopted for the sintered body form important factors. By the sintering operation in air which has been adopted in the ordinary sintering, the coloration attained at all is only in a light gray color or a blue color. Only by forming a sintered body in a non-oxidizing atmosphere at a temperature in the range of  $1200 - 1500^\circ\text{C}$ , it is made possible to have the produced sintered body to assume a black tone. As regards the mechanism responsible for the assumption of the black tone in such an atmosphere at such a temperature as specified above, it is inferred that the lack of oxygen likewise constitutes itself a factor of some sort or other.

[0030]

For the sintering atmosphere, such a non-oxidizing gas as argon, helium, nitrogen, and hydrogen may be used. The sintering may be performed in a reducing atmosphere formed by having a hydrogen gas partly incorporated in such an inert gas as argon, which results in enhancing the impartation of

a black tone.

[0031]

As respects the sintering temperature, if it is less than 1200°C, the shortage would be at a disadvantage in rendering it difficult to obtain a fully densified sintered body and, even when the sintered body is densified at all, preventing this sintered body from thoroughly undergoing coloration in black. If the temperature exceeds 1500°C, the excess would be at a disadvantage in suffering the crystal phase of a produced sintered body to melt and disrupting any plan to obtain a normal sintered body on account of such phenomena as fusion and expansion. The sintering temperature is more preferably in the range of 1275°C - 1450°C.

[0032]

Regarding the thermal expansion coefficient of the sintered body of this invention, the thermal expansion coefficient is required to be not more than  $0.6 \times 10^{-6}/^{\circ}\text{C}$  in absolute value at room temperature in the light of the necessity for maintaining the dimensional accuracy and the stability needed in production units for the recent high integration semiconductors and miniaturized magnetic heads. The precision members which demand thermal stability of still higher accuracy are in need of thermal expansion coefficients approximating closely to zero expansion. The thermal expansion coefficient is preferred to be not more than  $0.3 \times 10^{-6}/^{\circ}\text{C}$  (namely  $-0.3 - 0.3 \times 10^{-6}/^{\circ}\text{C}$ ) in absolute value at room temperature. Here, the minus numerical value in the thermal expansion coefficient means the fact that the relevant member shrinks as the temperature rises. In the narrow temperature range around room temperature, a sintered body having minus thermal expansion coefficient while having the composition conforming to this invention may be obtained.

[0033]

Concerning the rigidity (Young's modulus) of a sintered body, in order for the sintered body to serve effectively as a precision structure within a fixed space, the rigidity is required to have a Young's modulus of not less than 100 GPa, optimally of not less than 120 GPa. If Young's modulus is less than 100 GPa, the shortage would be at a disadvantage in requiring the structure to increase in wall thickness and size with a view to repressing the deformation of the relevant member.

[0034]

Where a given sintered body is to be used in such a partial supporting member as an end face supporting shaft, the specific rigidity (Young's modulus/specific gravity) must be also high in order for the sintered body to retain precision fully sufficient for a structure. In this invention, the specific rigidity is required to be not less than 40 GPa·cm<sup>3</sup>/g, preferably not less than 50 GPa·cm<sup>3</sup>/g.

[0035]

In the sintered body of this invention, by being provided with a chemical composition mainly comprising 8.0 - 17.2 mass % of MgO, 22.0 - 38.0 mass % of Al<sub>2</sub>O<sub>3</sub>, 49.5 - 65.0 mass % of SiO<sub>2</sub>, a crystal phase of the sintered body can be formed as a crystal phase made mainly of cordierite and the thermal expansion coefficient in absolute value at room temperature, the modulus of elasticity (Young's modulus), and the specific rigidity (Young's modulus/specific gravity) can be adjusted within the respective ranges contemplated by this invention. Though Li<sub>2</sub>O is not an essential component for the sintered body, the sintering properties may be enhanced and the impartation of a black tone may be promoted by the incorporation of Li<sub>2</sub>O.



[0036]

The chemical composition of MgO, Al<sub>2</sub>O<sub>3</sub>, and SiO<sub>2</sub> is so fixed as to fall in the percentage composition as mentioned above with a view to enabling the sintered body to acquire  
5 a crystal phase formed mainly of cordierite. If the proportions of MgO and Al<sub>2</sub>O<sub>3</sub> are unduly large, the excess would be at a disadvantage in suffering the crystal phase of spinel, mullite, or forsterite to acquire an unduly large thermal expansion coefficient and the thermal expansion coefficient  
10 of the sintered body as a whole to exceed  $0.6 \times 10^{-6} / ^\circ\text{C}$ . Conversely, if the proportions of MgO and Al<sub>2</sub>O<sub>3</sub> are unduly small, the shortage would be at a disadvantage in unduly decreasing the crystal phase of cordierite. If the proportion of SiO<sub>2</sub> is unduly large, the excess would induce a decrease  
15 in the modulus of elasticity. If this proportion is unduly small, the shortage would unduly increase such crystal as spinel, mullite, or forsterite which has high thermal expansion coefficient.

[0037]

20 The sintered body of this invention is enabled to be improved in the sintering properties by incorporating therein Li<sub>2</sub>O in addition to MgO, Al<sub>2</sub>O<sub>3</sub>, and SiO<sub>2</sub>. This improvement results in facilitating the formation of a densified sintered body. The start of the activation of the reaction (mass  
25 transfer) in the sintered body from a relatively low temperature onward can bring about such effects as enabling the impartation of a black color to start at a low temperature and the finally obtained sintered body to assume a black tone to a greater extent. The addition of Li<sub>2</sub>O in any amount  
30 exceeding 2.5 mass % is unfavorable because the excess would bring about a conspicuous decrease in the modulus of elasticity. For the purpose of deriving expected effects from the addition

of  $\text{Li}_2\text{O}$ , this compound is preferred to be added in an amount of not less than 0.1 mass %. By adjusting the amount of  $\text{Li}_2\text{O}$  to be added in the range of 0.2 - 1.0 mass %, it is made possible to obtain a sintered body having such an extremely low thermal expansion coefficient as of not more than  $0.1 \times 10^{-6} / ^\circ\text{C}$  in absolute value.

[0038]

In this invention, for the purpose of acquiring the satisfactory low thermal expansion coefficient and rigidity, the crystal phase of cordierite in the sintered body is preferred to account for not less than 80 vol. %, optically not less than 90 vol. %, of the whole volume of the sintered body. The expression "crystal phase of cordierite" as used in this invention means a crystal phase which comprises pure cordierite crystal plus a crystal phase which, on X ray diffraction, manifests a diffraction peak of cordierite and yet reveals a change in the lattice constant owing to a solid solution of Li and transition elements.

[0039]

The crystal phase of the sintered body of this invention may be a single-phase crystal phase of cordierite or may additionally incorporate therein a  $\text{Li}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2$  type crystal phase ( $\beta$ -spodumene, eucryptite, petalite). Any crystal phase other than these crystal phases is preferred not to be contained in the sintered body from the viewpoint of acquiring a low thermal expansion coefficient, though the incorporation thereof can be tolerated when the content thereof is not more than 5 mass %.

[0040]

Of the sintering method used for the production of the sintered body of this invention, such elements as the sintering atmosphere and the sintering temperature have been already

described above.

[0041]

The sintering method itself may be selected among a hot  
press method, a hot isostatic press (HIP) method, a gas pressure  
5 sintering method, and a normal pressure sintering method.  
The hot press method, the HIP method, and the gas pressure  
sintering method prove particularly favorable in a sense that  
they prevent low melting substances from foaming and enable  
the sintering process to perform at a higher temperature and  
10 they permit a sintered body to be formed within a percentage  
composition in which the normal pressure sintering method  
fails to produce required sintering. Economically, the  
normal pressure sintering method excels all the other methods  
enumerated above and can be applied to members complicate  
15 and large. Thus, the sintering method can be used as varied  
with the kind of application to be adopted.

[0042]

Since the HIP method and the hot press method are capable  
of producing poreless materials (materials of specular  
20 surfaces), they can be applied to the production of materials  
for reflecting mirrors, fastener materials which abhor the  
deposition of dirt in pores, and transportation grade band  
members.

[0043]

25 Since the sintered body of this invention is intended  
for such uses as cherish precision, it is not allowed to change  
size and geometric precision over time and to generate an  
outer gas. The sintered body, therefore, is required to have  
a dense texture. For this reason, the apparent porosity is  
30 required to be not more than 2%, preferably not more than  
0.2%. Even for the sake of securing the rigidity contemplated  
by this invention, it is necessary that the apparent porosity

be kept to a level of not more than 2%. By adopting the method for the production of the sintered body of this invention which has been described so far, it is made possible to adjust the apparent porosity at a value in the range of this invention  
5 specified above.

[0044]

The following materials can be used as the raw material powder to be used for the production of the sintered body of this invention.

10 [0045]

Electro-molten cordierite, synthetic cordierite, talc, magnesia, magnesium hydroxide, magnesium carbonate, and magnesia spinel are usable as MgO sources, silica is usable as a SiO<sub>2</sub> source, alumina is usable as an Al<sub>2</sub>O<sub>3</sub> source, and  
15 kaolin and mullite are usable as an Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> composite source.

[0046]

The electro-molten cordierite and synthetic cordierite may function also as a SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub> source, the magnesia spinel may function also as an Al<sub>2</sub>O<sub>3</sub> source, and the talc may function  
20 also as a SiO<sub>2</sub> source.

[0047]

As the raw material for the MgO source, it is advantageous to supply not less than 70 mass % of the total MgO component of the sintered body with electro-molten cordierite,  
25 synthetic cordierite, or talc. When the proportion occupied by this raw material is set at not less than 70 mass % of the total MgO component, the impartation of a black color can be improved to a greater extent. Though the mechanism responsible for inducing this phenomenon has not been  
30 elucidated fully, it may be logically explained by a supposition that the difference in the crystal phase of the sintered body occurring from the intermediate phase through

the terminal phase of the sintering process produces an influence.

[0048]

Also from the standpoint of improving the yield of  
5 sintering of articles in a large shape or in a complicated  
shape, the use of the raw materials of electro-molten  
cordierite, synthetic cordierite, and talc prove  
advantageous.

[0049]

10 As  $\text{Li}_2\text{O}$  sources, lithium carbonate, lithium oxide,  
 $\beta$ -spodumene, eucryptite, and petalite powder can be used.  
From the viewpoint of improving sintering properties of  
products of a large shape and a complicate shape, the  
 $\beta$ -spodumene, eucryptite, and petalite powder are particularly  
15 suitable as raw material powders.

[0050]

As transition element sources, oxides, hydroxides,  
nitrates, and carbonates of transition elements, and the metal  
powders thereof can be used.

20 [0051]

[Examples]

As raw material powders, magnesia (average particle  
diameter 0.2  $\mu\text{m}$ ), talc (average particle diameter 5  $\mu\text{m}$ ),  
electro-molten cordierite (average particle diameter 3  $\mu\text{m}$ ),  
25 synthetic cordierite (average particle diameter 2.5  $\mu\text{m}$ ),  
magnesium hydroxide (average particle diameter 0.5  $\mu\text{m}$ ),  
magnesium carbonate (average particle diameter 1  $\mu\text{m}$ ), lithium  
carbonate (average particle diameter 2  $\mu\text{m}$ ), lithium oxide  
(average particle diameter 1  $\mu\text{m}$ ),  $\beta$ -spodumene (average  
30 particle diameter 5  $\mu\text{m}$ ), eucryptite (average particle diameter  
5  $\mu\text{m}$ ), petalite (average particle diameter 4  $\mu\text{m}$ ), silica  
(molten silica, average particle diameter 0.7  $\mu\text{m}$ ), alumina

(average particle diameter 0.3  $\mu\text{m}$ ), kaolin (average particle diameter 2.5  $\mu\text{m}$ ), and mullite (average particle diameter 1  $\mu\text{m}$ ) were used. The synthetic cordierite was obtained by mixing magnesia, silica, and alumina powder at a ratio satisfying a theoretical composition of cordierite crystal ( $2\text{MgO} \cdot 2\text{Al}_2\text{O}_3 \cdot 5\text{SiO}_2$ ), and allowing the components to react with one another at 1420°C for 10 hours thereby transforming the mixture into a cordierite in a granular form. The granular cordierite was pulverized prior to use.

10 [0052]

As the raw materials for the transition element sources, oxides, hydroxides, nitrates, carbonates, or metal powders of the relevant transition elements were used.

[0053]

15 [Table 1]

Table 1

No.	Raw material powder to used	Chemical composition (mass %)						
		MgO	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	Li <sub>2</sub> O	X(*1)	Y(*2)	Coloring auxiliary (*3)
1	Talc · petalite Mullite · silica	13.1	35.0	51.1	0.3	3.71	1.38	Fe <sub>2</sub> O <sub>3</sub> 0.3% CoO 0.2%
2	Electro-molten cordierite · silica Euciptite · mullite	9.8	28.4	60.4	0.6	5.67	1.96	Fe <sub>2</sub> O <sub>3</sub> 0.6% Cr <sub>2</sub> O <sub>3</sub> 0.2%
3	Talc · β-spodumene Kaolin · alumina	10.6	36.2	52.4	0.1	4.87	1.43	Fe <sub>2</sub> O <sub>3</sub> 0.7%
4	Electro-molten cordierite · silica petalite · alumina	12.6	32.3	54.5	0.4	4.07	1.59	Fe <sub>3</sub> O <sub>4</sub> 0.2%
5	Talc · kaolin · alumina Lithium carbonate · silica	15.8	22.4	60.8	0.2	3.72	2.64	Nickel sulfate 0.4% Cr <sub>2</sub> O <sub>3</sub> 0.3%
6	Synthetic cordierite · silica Lithium hydroxide · mullite	9.8	29.5	57.5	2.0	4.23	1.41	Fe <sub>2</sub> O <sub>3</sub> 1.0% Cr <sub>2</sub> O <sub>3</sub> 0.2%
7	Talc · β-spodumene Alumina · silica · magnesia	11.5	33.4	54.2	0.7	4.23	1.46	Fe <sub>2</sub> O <sub>3</sub> 0.2%
8	Electro-molten cordierite · silica petalite · alumina	8.2	29.7	60.2	1.4	5.98	1.65	Copper oxide 0.3% Manganese carbonate 0.2%

\*1 X = (SiO<sub>2</sub> - 8 × Li<sub>2</sub>O)/MgO (mass ratio)\*2 Y = (SiO<sub>2</sub> - 8 × Li<sub>2</sub>O)/Al<sub>2</sub>O<sub>3</sub> (mass ratio)

\*3 The amount of a given coloring auxiliary is expressed in mass % as reduced to the relevant oxide.

Table 1 (Continued)

No.	Raw material powder to used	Chemical composition (mass %)					
		MgO	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	Li <sub>2</sub> O	X(*1)	Y(*2)
9	Electro-molten cordierite · silica Eucryptite · mullite	11.3	30.3	57.9	0.2	4.98	1.86
10	Talc · petalite · silica Magnesium hydroxide · alumina	13.0	33.4	51.7	0.4	3.73	1.45
11	Electro-molten cordierite · silica β-spodumene · alumina	12.3	33.0	53.7	0.6	3.98	1.48
12	Talc · magnesia Lithium · silica · kaolin	13.8	24.2	60.2	0.8	3.90	2.22
13	Electro-molten cordierite · silica Lithium oxide · alumina	15.2	32.0	51.9	0.1	3.36	1.60
14	Talc · β-spodumene Alumina · silica	9.9	30.1	58.4	1.0	5.09	1.67
15	Synthetic cordierite · silica β-spodumene · alumina	8.5	28.9	60.4	1.2	5.98	1.76
16	Electro-molten cordierite · silica Alumina · magnesium hydroxide	10.7	34.5	54.3	0.0	5.07	1.57

\*1 X = (SiO<sub>2</sub> - 8 × Li<sub>2</sub>O)/MgO (mass ratio)\*2 Y = (SiO<sub>2</sub> - 8 × Li<sub>2</sub>O)/Al<sub>2</sub>O<sub>3</sub> (mass ratio)

\*3 The amount of a given coloring auxiliary is expressed in mass % as reduced to the relevant oxide.



Table 1 (Continued)

No.	Raw material powder to used	Chemical composition (mass %)					
		MgO	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	Li <sub>2</sub> O X(*1)	Y(*2)	Coloring auxiliary (*3)
17	Talc · alumina · silica β-spodumene · mullite	14.8	36.2	48.2	0.5	2.99	Fe <sub>2</sub> O <sub>3</sub> 0.3%
18	Electro-molten cordierite · talc Silica · lithium oxide · alumina	18.5	26.3	54.3	0.2	2.85	Fe <sub>2</sub> O <sub>3</sub> 0.7%
19	Talc · kaolin · silica Lithium oxide · alumina	16.4	20.9	61.8	0.1	3.72	Copper oxide 0.5% Cr <sub>2</sub> O <sub>3</sub> 0.7%
20	Electro-molten cordierite · silica Lithium oxide · mullite	12.5	34.8	52.3	0.4	3.93	None
21	Synthetic cordierite · silica β-spodumene · alumina	8.5	28.9	60.4	1.2	5.98	Fe <sub>2</sub> O <sub>3</sub> 1.0%
22	Talc · petalite Lithium carbonate · alumina	9.3	31.8	56.5	2.4	4.01	None
23	Talc · kaolin β-spodumene	5.7	30.6	58.4	5.3	2.81	None

\*1 X = (SiO<sub>2</sub> - 8 × Li<sub>2</sub>O)/MgO (mass ratio)\*2 Y = (SiO<sub>2</sub> - 8 × Li<sub>2</sub>O)/Al<sub>2</sub>O<sub>3</sub> (mass ratio)

\*3 The amount of a given coloring auxiliary is expressed in mass % as reduced to the relevant oxide.

[0054]  
[Table 2]

Table 2

Example of This Invention								
No.	Sintering method sintering temperature	Gas for atmosphere	Thermal expansion coefficient at room temperature 10 <sup>-6</sup> /°C	Apparent porosity %	Young's modulus GPa	Specific rigidity GPa·cm <sup>3</sup> /g	Total reflectivity %	Cordierite crystal phase ratio %
1	Hot press, 1420°C	Argon + hydrogen 5%	0.14	0	141	52.2	9.8	100
2	Gas pressure, 1400°C	Argon	0.11	0	112	48.2	11.2	100
3	HIP, 1375°C	Argon	0.21	0	157	58.8	9.2	96
4	Normal pressure, 1370°C	Argon	0.01	0.1	134	54.2	11.8	100
5	Hot press, 1420°C	Argon + hydrogen 5%	0.35	0	121	51.5	14.5	91
6	Gas pressure, 1420°C	Argon	-0.09	0	120	46.5	15.2	96
7	Normal pressure, 1390°C	Argon + hydrogen 5%	0.14	0.2	129	50.6	12.2	100
8	Gas pressure, 1290°C	Argon	-0.18	0	108	48.0	15.2	97

Table 2 (Continued)

Example of This Invention								
No.	Sintering method sintering temperature	Gas for atmosphere	Thermal expansion coefficient at room temperature 10 <sup>-6</sup> /°C	Apparent porosity %	Young's modulus GPa	Specific rigidity GPa·cm <sup>3</sup> /g	Total reflectivity %	Cordierite crystal phase ratio %
9	Normal pressure, 1385°C	Helium	0.04	0.1	147	59.8	11.2	100
10	Hot press, 1420°C	Argon + hydrogen 5%	0.05	0	138	53.2	8.9	100
11	Normal pressure, 1350°C	Argon	-0.01	0.1	128	54.4	10.9	100
12	Hot press, 1320°C	Argon + hydrogen 5%	0.28	0	118	49.8	9.7	97
13	Gas pressure, 1450°C	Argon	0.52	0	185	68.5	10.2	86
14	Hot press, 1250°C	Argon	0.18	0	121	50.0	16.1	100
15	Normal pressure, 1300°C	Argon + hydrogen 5%	0.25	0.1	120	52.0	7.9	98
16	Hot press, 1420°C	Argon + hydrogen 5%	0.29	0	151	56.5	14.6	100

Table 2 (Continued)

Comparative Example									
No.	Sintering method Sintering temperature	Gas for atmosphere	Thermal expansion coefficient at room temperature $10^{-6}/^{\circ}\text{C}$	Apparent porosity %	Young's modulus GPa	Specific rigidity $\text{GPa}\cdot\text{cm}^3/\text{g}$	Total reflectivity %	Cordierite crystal phase ratio %	
17	Hot press, $1400^{\circ}\text{C}$	Argon + hydrogen 5%	1.95	0	141	50.2	38.2	89	
18	Gas pressure, $1450^{\circ}\text{C}$	Argon	1.42	0	145	55.0	19.3	76	
19	Normal pressure, $1300^{\circ}\text{C}$	Argon	0.98	0.4	82	40.0	18.5	72	
20	Normal pressure, $1390^{\circ}\text{C}$	Argon	0.15	0.1	117	46.6	29.7	97	
21	Normal pressure, $1180^{\circ}\text{C}$	Argon + hydrogen 5%	0.42	5.1	72	36.2	43.5	54	
22	Normal pressure, $1370^{\circ}\text{C}$	Air	0.52	0.9	88	39.0	80.2	55	
23	Normal pressure, $1280^{\circ}\text{C}$	Air	0.60	2.8	65	32.0	74.6	46	

[0055]

As shown in Table 1, the samples of Example Nos. 1 - 16 and Comparative Example Nos. 17 - 23 were obtained by combining the relevant raw material powders in such proportions as form the chemical compositions shown in Table 1, adding 3 mass parts of a resin binder to each of the resultant mixtures, and mixing the produced blends each with water as a solvent in an alumina pot mill for 24 hours. The resultant slurries were each dried and granulated and then formed in a prescribed shape under static pressure of  $1500 \text{ kg/cm}^2$  ( $147 \text{ MPa}$ ). The formed product was heated in air to  $500^\circ\text{C}$  to degrease the resin binder.

[0056]

The degreased formed product was sintered by a sintering method, in a sintering atmosphere, at a sintering temperature mentioned in Table 2. The sintering was performed under a surface pressure of  $400 \text{ MPa}$  by the hot press method, under a gas pressure of  $50 \text{ kg/cm}^2$  ( $5 \text{ MPa}$ ) by the gas pressure sintering method, and under 1500 atmospheres at  $1300^\circ\text{C}$  after the step of normal pressure sintering by the HIP method. The sintering times each at a relevant temperature indicated in the tables were 4 hours in the normal pressure sintering and the gas pressure sintering, and 1 hour in the hot press sintering method and the HIP sintering method.

[0057]

The produced sintered bodies were each tested for thermal expansion coefficient at room temperature, total reflectivity, apparent porosity, Young's modulus, specific rigidity, and cordierite crystal phase ratio. The results are shown in Table 2. The total reflectivity was determined in accordance with JIS K7105. Since the thermal expansion coefficient at room temperature requires accurate determination, the

determination was carried out in accordance with JIS R3251 (dual light path Michelson's laser interference method) for the use in the determination of a thermal expansion coefficient of a low thermal expansion glass. The apparent porosity was  
5 determined by the Archimedes method. The cordierite crystal phase ratio was determined by the X-ray diffraction, on the condition that the crystal phase comprising pure cordierite crystals and a crystal phase having a lattice constant altered by a solid solution of Li and a transition element while  
10 possessing a diffraction peak of cordierite calculated was reckoned as a cordierite crystal phase.

[0058]

Experiment Nos. 1 - 16 represented working examples of this invention, which invariably produced satisfactory  
15 results.

[0059]

Experiment Nos. 17 - 23 represented comparative examples.

The sample of No. 17 showed thermal expansion coefficient and total reflectivity both outside the respective ranges contemplated by the invention because the  $\text{SiO}_2$  composition and the ratio X thereof both deviated from the ranges of this  
20 invention.

The sample of No. 18 showed thermal expansion coefficient, total reflectivity, and cordierite crystal phase ratio all outside the respective ranges contemplated by the invention because the MgO composition and the ratio X both deviated from the ranges of this invention.  
25

The sample of No. 19 showed thermal expansion coefficient, Young's modulus, total reflectivity, and cordierite crystal phase ratio all outside the respective ranges contemplated by this invention because the  $\text{Al}_2\text{O}_3$  composition deviated from  
30

the range of this invention.

The sample of No. 20 showed total reflectivity outside the relevant range contemplated by this invention because it had no transition elements added.

5       The sample of No. 21 showed apparent porosity, Young's modulus, specific rigidity, total reflectivity, and cordierite crystal phase ratio invariably outside the respective ranges contemplated by this invention because the sintering temperature was below the lower limit specified  
10 by this invention.

      The sample of No. 22 showed the ratio Y outside the range fixed by this invention and was sintered in air without any addition of a transition element. Specifically, the method disclosed in the publication of JP-A-61-72,679 was performed  
15 on this sample. As a result, this sample showed total reflectivity widely deviating from the range fixed by this invention and Young's modulus, specific rigidity, and cordierite crystal phase ratio also falling outside the respective ranges contemplated by this invention.

20       The sample of No. 23 showed MgO and Li<sub>2</sub>O compositions and ratios X and Y outside the respective ranges fixed by this invention and was sintered in air without adding any transition element. Specifically, the method disclosed in the publication of JP-A-10-53,460 was performed on this sample.  
25 As a result, the total reflectivity of the sample widely deviated from the range fixed by this invention and apparent porosity, Young's modulus, specific rigidity, and cordierite crystal phase ratio thereof were outside the respective ranges contemplated by this invention.

30       [0060]

      [Effects of the Invention]

      By this invention, it is made possible to realize a black



low thermal expansion ceramic sintered body having a black  
tone, manifesting very low thermal expansion at room  
temperature, and abounding in rigidity and specific rigidity  
and a method for the production thereof. For the first time,  
5 therefore, this invention has materialized the qualities that  
have been demanded by such members requiring a black tone  
in devices using laser beam and ultraiviolet light while  
securing dimensional accuracy and stability necessary for  
the production units for recent highly integrated  
10 semiconductors and miniaturized magnetic heads.

[DOCUMENT'S NAME] ABSTRACT

[ABSTRACT]

[Problem] To provide a black low thermal expansion ceramic sintered body having a black tone, manifesting very small  
5 thermal expansion at room temperature and abounding in rigidity and specific rigidity, and a method for the production thereof.

[Solving Means] A black low thermal expansion high specific rigidity ceramic sintered body characterized by  
10 having a chemical composition comprising 8.0 - 17.2 mass % of MgO, 22.0 - 38.0 mass % of  $\text{Al}_2\text{O}_3$ , 49.5 - 65.0 mass % of  $\text{SiO}_2$ , a total of 0.1 - 2 mass % of one or more transition elements as reduced to oxides, and 0 - 2.5 mass % of  $\text{Li}_2\text{O}$ , and having the mass ratios satisfy the relationships of  $(\text{SiO}_2 - 8 \times$   
15  $\text{Li}_2\text{O})/\text{MgO} \geq 3.0$  and  $(\text{SiO}_2 - 8 \times \text{Li}_2\text{O})/\text{Al}_2\text{O}_3 \geq 1.2$ .

[Selected Drawing] None